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Evaluation and modelling of the size fractionated aerosol particle number concentration measurements nearby a major road in Helsinki – Part I: Modelling results within the LIPIKA project

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Abstract. A field measurement campaign was conducted near a major road “Itäväylä” in an urban area in Helsinki in 17–20 February 2003. Aerosol measurements were conducted using a mobile laboratory “Sniffer” at various distances from the road, and at an urban background location. Measurements included particle size distribution in the size range of 7 nm–10 μ m (aerodynamic diameter) by the Electrical Low Pressure Impactor (ELPI) and in the size range of 3–50 nm (mobility diameter) by Scanning Mobility Particle Sizer (SMPS), total number concentration of particles larger than 3 nm detected by an ultrafine condensation particle counter (UCPC), temperature, relative humidity, wind speed and direction, driving route of the mobile laboratory, and traffic density on the studied road. In this study, we have compared measured concentration data with the predictions of the road network dispersion model CAR-FMI used in combination with an aerosol process model MONO32. For model comparison purposes, one of the cases was additionally computed using the aerosol process model UHMA, combined with the CAR-FMI model. The vehicular exhaust emissions, and atmospheric dispersion and transformation of fine and ultrafine particles was evaluated within the distance scale of 200 m (corresponding to a time scale of a couple of minutes). We computed the temporal evolution of the number concentrations, size distributions and chemical compositions of various particle size classes. The atmospheric dilution rate of particles is obtained from the roadside dispersion model CAR-FMI. Considering the evo-

lution of total number concentration, dilution was shown to be the most important process. The influence of coagulation and condensation on the number concentrations of particle size modes was found to be negligible on this distance scale. Condensation was found to affect the evolution of particle diameter in the two smallest particle modes. The assumed value of the concentration of condensable organic vapour of 10^{12} molecules cm^{-3} was shown to be in a disagreement with the measured particle size evolution, while the modelling runs with the concentration of condensable organic vapour of 10^9 – 10^{10} molecules cm^{-3} resulted in particle sizes that were closest to the measured values.

1 Introduction

Elevated particle concentrations have been associated with short and long term health effects (e.g. WHO, 2004). There is also evidence that the ultrafine particle fraction is associated with deterioration of respiratory health (Peters et al., 1997). Therefore high number concentrations of fine and ultrafine particles in urban environments, especially in the vicinity of major streets and roads, have raised the interest to study the physical and chemical transformation of PM in the urban environment. This work is a continuation of our effort to analyse particle transformation and dilution in a local scale. In our earlier work, we have studied numerically the transformation and dilution in distance scale of less than 100 m from a road with a combination of the aerosol process model MONO32 and a simple plume model (Pohjola et al., 2003, 2004). Dilution was found to be the most important

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process affecting the particle number concentrations, and effect of condensation was seen to depend on the available concentration of condensable organic vapour. However, only a small set of experimental data was available for the evaluation of the numerical computations.

Of the aerosol processes, nucleation was not considered in this work. According to the theoretical work of Zhang and Wexler (2004) the first stage of dilution during 1–3 s would be accompanied with nucleation. At least for diesel exhaust nucleation is likely (Maricq et al., 2002; Zhang and Wexler, 2004). The modelling of nucleation would require more detailed information about the environmental conditions very near the tailpipe (e.g. temperature gradient, information about chemical composition and concentrations of volatile nucleating vapours), which was not available. We also did not assess the influence of dry deposition, as it was previously shown to be irrelevant on this time scale (Ketzel and Berkowicz, 2004).

Dispersion of particle number concentrations and size distributions at various distances from roads have been measured by several groups, e.g. in the United States (Zhu et al., 2002a, b, 2004), in Australia (Morawska et al., 1999; Hitchins et al., 2000; Gramotnev and Ristovski, 2004), in United Kingdom (Shi et al., 1999, 2001) and in Finland (Pirjola et al., 2006). In most of those studies the measurements were taken at 15 m and further up to 300 m downwind from the highway. However, in the last mentioned case, during the Finnish LIPIKA campaign, a mobile laboratory was available, and the dispersion measurements were conducted directly on a highway and downwind to around a distance of 150 m. These measurements therefore provide a comprehensive data set for model evaluation.

The evolution of particle number concentrations and size distributions as a function of distance from a road or street, and the importance of aerosol processes has recently been analysed via the concept of aerosol process time scales by Zhang and Wexler (2004) and Ketzel and Berkowicz (2004), and with aerosol dynamics model simulations by Jacobson and Seinfeld (2004). In conditions, where dilution with cleaner background air is restricted and the exhausts accumulate in the available air space, as e.g. in a road tunnel (Gidhagen et al., 2003) or during a temperature inversion (Kerminen et al., 2007) the aerosol processes of coagulation, condensation and evaporation, have been shown to be important.

In this paper, modelling is the main issue, as the measurement dataset has already been presented and examined elsewhere (Pirjola et al., 2006). The main aims of this study were to model the evolution of particle number concentrations and size distributions as a function of distance from a road and to compare the predicted results with the data that has been measured in the national LIPIKA project. More detailed objectives were to study the importance of the concentration of volatile condensable vapour for this evolution, and to determine, which of the included aerosol processes (dilution and mixing with ambient air, coagulation and condensa-

tion) are important in the studied distance scale (the modelled distances extend up to 350 m). Compared to the previous work of the present authors in this area (Pohjola et al., 2003, 2004), the modelling of atmospheric dilution has been substantially improved, and the modelling also contains proper atmospheric boundary layer scaling (using the roadside dispersion model CAR-FMI combined with the meteorological pre-processing model MPP-FMI). The experimental dataset that is utilised in the evaluation of the predicted results is also substantially more extensive both in terms of the number of data and the measured parameters.

A companion paper (Part II) focuses, instead of modelling, on the analysis and evaluation of measurement data that has been obtained in the EU-funded SAPPHERE project. That paper discusses a comparison of the data measured at one roadside location with the corresponding data measured at an urban background location. In particular, the companion paper contains an evaluation of the temporal variations and dependencies on local meteorological conditions of the measured particle number concentrations and size distributions.

The emission estimates (including both emission factors and sizewise chemical composition) presented in this study (Part I) have been used in the modelling work presented in the companion paper (Part II). This paper (Part I) also presents the source term estimation methods and the offline coupling method of the dispersion model and aerosol process models; these mathematical methods will subsequently be used in both of these papers.

2 Experimental

2.1 Measuring locations and equipment

In this work, we consider the aerosol measurements conducted by a mobile laboratory, Sniffer, at various locations near the highway Itäväylä, a major road in an urban area of Helsinki, during the LIPIKA campaign in 17–20 February 2003, weekdays from Monday to Thursday (Pirjola et al., 2006). A schematic description of the measurement site is presented in Fig. 1. The highway consists of six lanes, three lanes to both directions (the total width of three lanes is 12 m), and a 6 m wide central grass area between the lanes to both directions. The speed limit is 80 km h⁻¹ and the average vehicle speed in the vicinity of the measurement site was approximately 70 km h⁻¹.

The measurement site is located about 5 km northeast of the city centre in a suburban area with substantial small-scale industrial capacity. The closest factory building with the height of approximately 7 m (2 floors) is located in the next block north or northeast of the measurement sites. However, to south and southwest of the street, in which the measurements were conducted (Fig. 1) there are no buildings, only some individual deciduous trees of an average height of 4 m. The factory periodically emits particles which contribute to

the local background aerosol. However, the data presented in this paper include only north-westerly wind sectors, which allowed for the measurements of the traffic exhausts originated from the road of Itäväylä.

The urban background was measured at the site of Saunalahti bay that is located approximately at a distance of 600 m northwest of the road. There are no major pollution sources in the immediate vicinity of this station. The urban background measurements were conducted before and after the roadside measurements a few times each day: in the morning before and after the morning rush hours, in the afternoon, and in late afternoon after the rush hours, altogether 189 min. The time resolution of the urban background measurements was 10 min or better.

The mobile laboratory used was constructed into a Volkswagen LT35 diesel vehicle (Pirjola et al., 2004). Particle size distribution in the size range of 7 nm–10 μ m (aerodynamic diameter) with 12 channels is measured by the Electrical Low Pressure Impactor (ELPI, Dekati Ltd, Keskinen et al., 1992) with a time resolution of 1 s. The nucleation mode particle size distribution was measured with a high size resolution by a Hauke type Scanning Mobility Particle Sizer (SMPS), where particles are first neutralised, then classified by a Differential Mobility Analyser (DMA) based on their electrical mobility, and counted by a CPC 3025 (TSI, Inc.). Measurement size range was 3–50 nm (mobility diameter), with a resolution of 20 channels and a scan-up time mostly 30 s. Additionally, the total number concentration of particles larger than 3 nm is detected by an ultrafine condensation particle counter CPC 3025 (TSI, Inc.). A passive clean air dilution system (ratio 1:3) was installed, in order to keep inside the measuring range of the instrument. The particle measurement inlet is located at the height of 2.4 m.

The weather station is located at the roof of the van. Temperature and relative humidity (Model HMP45A, Vaisala) as well as relative (in case of a moving vehicle) wind speed and direction by an ultrasonic wind sensor (Model WAS425AH, Vaisala) were measured at the height of 2.9 m. Additionally, a global position system (GPS V, Garmin) detected the van speed and the driving route, and a video camera in the cab recorded visually the traffic situations.

Additional meteorological data were measured at a height of 10 m from the ground at the roof of a container that was located at a distance of 9 m from the edge of the road. Wind speed and direction data obtained from the container measurements were compared with the measurements by the mobile laboratory. Due to the disturbance of the flow caused by the measuring van itself and the surrounding buildings, especially at low altitudes, we decided to use the wind data measured at the roof of the container.

2.2 Traffic measurements

The traffic flow data at Itäväylä was recorded continuously using electronic loops installed below the street surface by

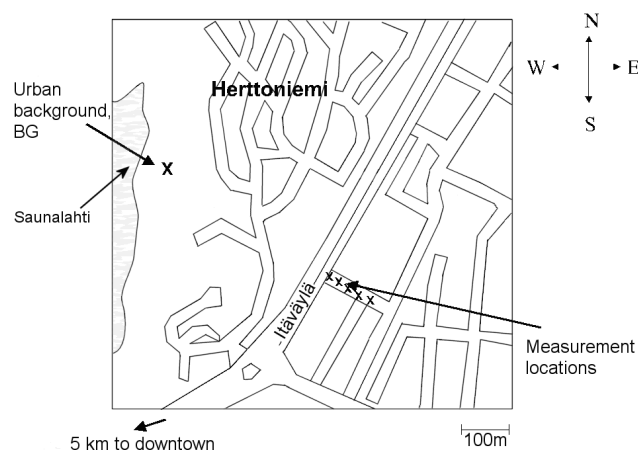


Fig. 1. Schematic map of the measurement site and its surroundings (Pirjola et al., 2006). The mobile laboratory measurements were conducted while the van was parked in several locations on a minor road (Työnjohtajankatu) that is approximately perpendicular to the major road studied (Itäväylä) and while driving on the major road.

the City of Helsinki and the Finnish Road Administration at a site located about 1.8 km southwest of the Itäväylä measurement site (Pirjola et al., 2006; Virtanen et al., 2006). However, there is one major junction between the traffic density count site and the measurement site. Additional manual three-minute vehicle counts (including the total traffic flow, and the fractions of light- and heavy-duty vehicles) were therefore performed at selected intervals at the mobile laboratory in order to check the representativity of the continuous traffic flow measurements. The agreement of the visual and the continuous electronic counts was excellent, indicating that the continuous measurements are well representative (Pirjola et al., 2006).

The measured traffic densities are presented in Fig. 2. The average daily traffic count during the time period considered was 46 063 vehicles per day, 50% of which was directed towards the city center on average. The average daily total traffic count varied between a maximum of 46 477 veh/day on 20 February and a minimum of 45 581 veh/day on 18 February, and the variations in the diurnal hourly traffic patterns were small within the four days studied. On all the days, the busiest traffic occurred during the morning and evening rush hours. The traffic during the morning hour contained on average about 8% of the daily traffic count (and 71% of the traffic during this hour was directed towards the city centre). The traffic at 04:00–05:00 p.m. contained on average about 9% of the total daily traffic count (and 64% of the traffic within this one hour was directed away from the centre).

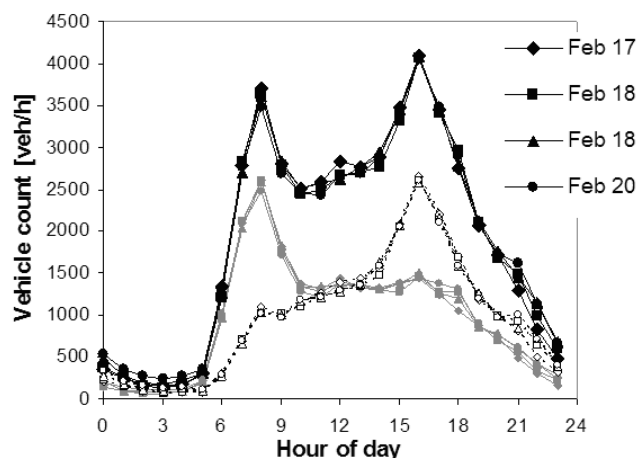


Fig. 2. The traffic densities measured at the traffic count site of Kulosaari for the days 17–20 February 2006. Total hourly traffic counts are indicated with the four bold black lines, traffic counts towards the city center are indicated with gray lines, and traffic counts directed away from the city are indicated with black dotted lines.

3 Modelling methods

3.1 Modelling of the vehicular emissions

Generally, particle number emission factors are not well known, as they depend on many factors related to variation of the vehicle fleet including fraction of diesel vehicles, driving speed and cycle, and ambient conditions such as ambient temperature, wind direction and relative humidity (Morawska et al., 2005; Ketzel et al., 2004).

In this study, emission factors for particles were calculated for the whole vehicle fleet characteristic of Itäväylä. In the Helsinki Metropolitan Area (HMA), light duty (LD) vehicles constitute about 90% of all traffic, 80% of which are petrol vehicles and 20% diesel vehicles (Kauhaniemi, 2003). It was assumed that all heavy duty (HD) vehicles are diesel operated. These percentiles were then used in calculating the emission factor for the total number concentration $EF(N_{\text{tot}})$ based on the corresponding LD ($\sim 1.1 \times 10^{14}$ particles $\text{veh}^{-1} \text{ km}^{-1}$) and HD ($\sim 5.2 \times 10^{15}$ particles $\text{veh}^{-1} \text{ km}^{-1}$) values, as reported by Gidhagen et al. (2004). As a result we obtained $EF(N_{\text{tot}}) = 6.23 \times 10^{14}$ particles $\text{veh}^{-1} \text{ km}^{-1}$.

To be able to estimate modal emission factors we have plotted the median size distribution of particles measured while driving on Itäväylä (Fig. 3). The diameters along with the lower and upper limits of the six modes were prescribed to vary according to this figure, in the range of $3 \text{ nm} < d < 7.5 \text{ nm}$ for the nucleation mode 1 (nuc1), $7.5 \text{ nm} < d < 43.2 \text{ nm}$ for the nucleation mode 2 (nuc2), $43.2 \text{ nm} < d < 0.122 \mu\text{m}$ for the Aitken mode (Ait), $0.122 \mu\text{m} < d < 0.321 \mu\text{m}$ and $0.321 \mu\text{m} < d < 2.5 \mu\text{m}$ for the accumulation modes 1 and 2 (acc1, acc2), respectively, and

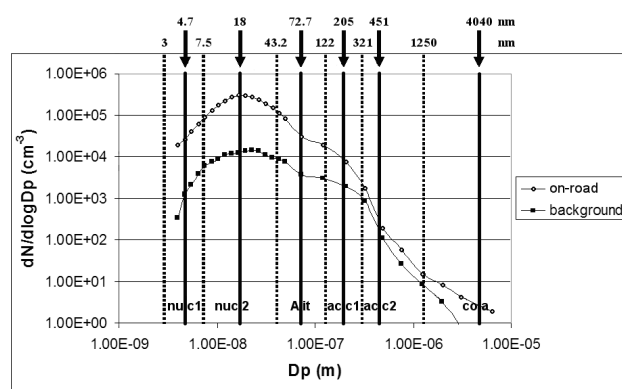


Fig. 3. Average size distribution of particles measured while driving on Itäväylä (open circles), and that for the median urban background (at 15:00–19:00) measured at the station of Saunalahti (black squares). The solid vertical lines show the assumed initial diameters of the six size modes used in the model, and the dashed vertical lines show the boundaries between these size modes. The numerical particle diameter values that correspond to these vertical lines have also been shown in the upper part of the figure.

$d > 2.5 \mu\text{m}$ for the coarse mode particles. The partitioning percents to each mode were calculated based on the measured total number concentration and concentrations of each size bin resulting in 9.5, 80.5, 8.7, 1.0, 0.3 and 0.001 percent, respectively. The modal emission factors were then obtained by multiplying $EF(N_{\text{tot}})$ by these numbers. Since the particle number concentration on Itäväylä is positively correlated with the traffic flow as seen in the Fig. 4, the emission factors also depend on the traffic flow. Thus we have finally extended the modal emission factors to allow for the temporal variation by multiplying them by the measured traffic density profiles, separately to both directions, to obtain Q_i in particles $\text{m}^{-1} \text{ s}^{-1}$.

For comparison, we also used the modal emission factors calculated based on the ELPI measurements by Sniffer. Yli-Tuomi et al. (2004) estimated the emission factor $EF(N_{\text{tot}})$ of 9.3×10^{15} particles per kg fuel for the highway conditions in HMA. A rough estimation of $EF(N_{\text{tot}}) = 8.7 \times 10^{14}$ particles $\text{veh}^{-1} \text{ km}^{-1}$ can be obtained by assuming that the average fuel consumption for light duty vehicles (90%) is 8 l/100 km and for heavy duty (10%) around 20 l/100 km. The results are discussed in Sect. 4.1.

For the modal chemical composition of the exhaust particles LIPIKA data for exhaust particle number concentrations measured while driving on the highway was used, combined with the chemical composition data extracted from Norbeck et al. (1998) (considering exhaust particle emissions for light duty vehicles) and Shi et al. (2000) (containing data on exhaust particle emissions for heavy duty vehicles). The properties of the particulate modes in the vehicle exhaust are presented in Table 1.

Table 1. The properties of the vehicle exhaust particle emissions.

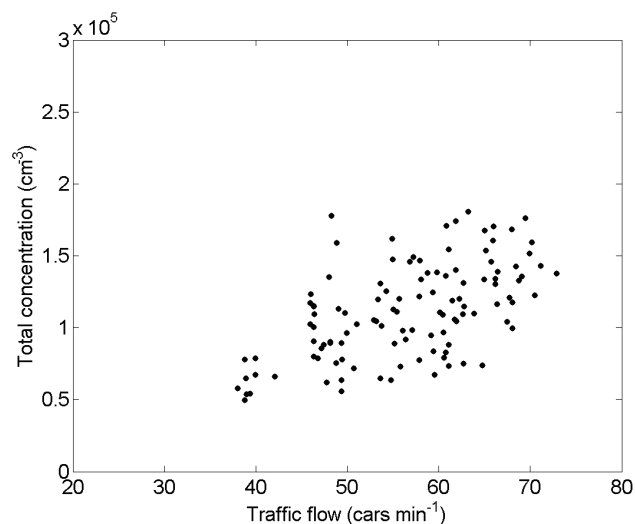
Particle size mode	Dry radius (nm)	% of total number concentration (cm^{-3})	Mass composition (%)						
			Sulphuric acid	Organic carbon	Elemental carbon	Mineral dust	Seasalt	Ammonium sulphate	Ammonium nitrate
Nucleation1	2.35	9.5	5	90	0	0	0	5	0
Nucleation2	9.0	80.5	5	90	0	0	0	5	0
Aitken	36.4	8.7	0	27	64	7	0	2.2	0.3
Accumulation1	103	1	0	26	63	9	0	3	0.3
Accumulation2	226	0.3	0	25	63	9	0	3	0.3
Coarse	1010	$9\text{e-}4\sim 0$	0	0	0	100	0	0	0

3.2 Evaluation of the particle size distribution and chemical composition of urban background

We utilized median urban background particle size distributions (one example presented in Fig. 3) that are based on the LIPIKA-measurements at the site of Saunalahti. The background size distributions were not measured simultaneously with the roadside concentrations. However, we have classified the measured time periods into three classes and calculated the medians over them; morning and afternoon rush hours at 07:00–09:00, at 15:00–19:00, and late evening at 21:00–23:00. The medians of the measured particle number concentrations varied between approximately 11 000–14 000 cm^{-3} ; the highest values were observed during the morning rush hour when more accumulation mode particles were present, and the lowest values during the night when also the nucleation mode diameter was somewhat smaller. As presented in Fig. 3 the median background particle size distribution has two modes, and the mode diameters do not differ significantly from the corresponding ones measured on-road. In this work the background to on-road –ratio of aerosol number concentration was lower for small particles and higher with larger particles; the particle number size distribution therefore is modified due to dilution and mixing.

The diameters of particles and the limits of the six modes were fitted to be the same as in the case of emission modelling (presented in Fig. 3).

The evaluation of the chemical composition of the background particles was based on Pakkanen et al. (2001a, b), concerning ultrafine particles and Viidanoja et al. (2002), concerning fine and coarse particles. The properties of the particulate modes in urban background air are presented in Table 2. The nucleation mode particles were assumed to be mainly composed of organic carbon, whereas the Aitken and accumulation mode particles included the most abundant components, organic and elemental carbon (OC and EC), in a ratio of OC/EC=1.2.

**Fig. 4.** Measured total particle number concentration at a distance of 9 m from the roadside as a function of traffic flow of the major road (Itäväylä).

3.3 Atmospheric dispersion and aerosol dynamics models

The CAR-FMI model includes an emission model, a dispersion model and statistical analysis of the computed time series of concentrations. The CAR-FMI model utilises the meteorological input data evaluated with the meteorological pre-processing model MPP-FMI. The dispersion equation is based on a semi-analytic solution of the Gaussian diffusion equation for a finite line source (Luhar and Patil, 1989). For a more detailed description of these models, the reader is referred to Härkönen et al. (2002) and Karppinen et al. (2000a, b). The performance of the CAR-FMI model has previously been extensively evaluated both against the results of field measurement campaigns regarding NO_x , NO_2 and O_3 (e.g. Kukkonen et al, 2001), $\text{PM}_{2.5}$ (Tiitta et al., 2002), and the data of urban concentration monitoring networks (e.g., Karppinen et al., 2000b; Kousa et al., 2001).

Table 2. The properties of particulate matter in the urban background air.

Particle size mode	Dry radius (nm)	Number concentration (cm ⁻³)	Total mass (ng m ⁻³)	Mass composition (%)						
				Sulphuric acid	Organic carbon	Elemental carbon	Mineral dust	Sea salt	Ammonium sulphate	Ammonium nitrate
Nucleation1	2.35	1.32e2	1.22e-2	7	68	19	0	0	0	6
Nucleation2	9.0	7.75e3	4.02e+1	7	68	19	0	0	0	6
Aitken	36.4	2.63e3	9.03e+2	21	24.5	19	12	2.5	0	21.3
Accumulation1	103	5.66e2	4.34e+3	21	24.5	19	12	2.5	0	21.3
Accumulation2	226	2.94e1	2.4e+3	21	24.5	19	12	2.5	0	21.3
Coarse	1010	1.7e-1	1.0e+4	2	23.4	4.56	59	7	0	4
Total		1.11e4	1.77e+4	10.2	24	10.8	38.5	5	0	11.5

The aerosol dynamics model MONO32 is a box model, which includes gas-phase chemistry and aerosol dynamics, and can be applied under clear sky conditions. The model uses monodisperse representation for particle size distribution with an optional number of size modes. In this work we have used six modes, two nucleation modes, one Aitken mode, two accumulation modes and one coarse mode. All particles in a mode are characterised by the same size and the same composition. Particles can consist of soluble material such as sulphuric acid, ammonium sulphate, ammonium nitrate and sodium chloride, organic carbon which can be soluble, partly soluble or insoluble, and insoluble material like elemental carbon and mineral dust. Size and composition of particles in any class can change due to multicomponent condensation of sulphuric acid and organic vapours as well as due to coagulation between particles. For a more detailed description of the MONO32 model and its evaluation against measurement data, the reader is referred to Pirjola and Kulmala (2000), Pirjola et al. (2003) and Pohjola et al. (2003).

The aerosol dynamics model UHMA (Korhonen et al., 2004) uses a sectional representation of the particle size distribution and includes the same gas-phase and aerosol microphysical processes as MONO32. In this work, we have used 50 fixed logarithmically spaced sections over the simulated size range. Particles in each individual section are assumed to be internally mixed; however, the composition is allowed to vary between the sections.

In this paper we have used the monodisperse model MONO32 as the primary aerosol process model; this model was also used in our earlier work in the same area (Pohjola et al., 2003). In that paper we studied the effects of different aerosol dynamic processes (condensation, coagulation, dry deposition, dilution and mixing with the ambient air) when traffic particle emissions disperse in a local scale. A simple semi-empirical plume model was used, and comparison with measured data was accomplished using a very limited set of data. These issues have been improved in this paper.

Clearly, various categories of aerosol dynamic models have advantages and disadvantages. The monodisperse models such as MONO32 are computationally more efficient than sectional models such as AEROFOR (Pirjola, 1999), AEROFOR2 (Pirjola and Kulmala, 2001) and UHMA (Korhonen et al., 2004); however, for the simulations presented in this study, expanding only over a time scale of few minutes, computational requirements do not limit the use of either kind of model set-ups. The MONO32 model is not subject to numerical diffusion upon condensation which might be a problem in sectional models. However, numerical diffusion can be reduced in sectional models by increasing the number of size sections or allowing diameter to move inside the sections.

In this study, we have used both a monodisperse model (MONO32) and a sectional aerosol dynamic model (UHMA) for one case, in order to inter-compare these numerical results. The same subroutines for aerosol processes are used in both models. It is well-known that monodisperse models are not well suited for simulating intensive continuous nucleation, unless a new mode is created for allocating for fresh 1 nm particles. However, in this paper nucleation is not considered. In general, the size distributions can be better represented by sectional models.

3.4 Offline-type coupled modelling of the emissions, dispersion and aerosol processes

The offline-type coupling explained in this chapter concerns CAR-FMI and the monodisperse model MONO32; however, the same mathematical procedure was applied in coupling CAR-FMI and the sectional model UHMA.

The dispersion model (CAR-FMI) is a three-dimensional second-generation (including boundary layer scaling) Gaussian model with a temporal resolution of one hour, i.e., the emissions, background concentrations and meteorological conditions are assumed to be constant during that period. The aerosol dynamics models used are zero-dimensional

Lagrangian models; these models address the temporal evolution of aerosols and gases within a constant volume of air. The temporal resolution can be selected, but it is commonly of the order of seconds. The combination of these two models in order to form an integrated system is therefore not a trivial task. However, the principle of combining the two models is simple: the dispersion model should provide for the dilution of the Lagrangian air parcel, and the aerosol process model should then allow for the aerosol transformation within this temporally expanding volume.

There is no analytical solution for combining the equations of the CAR-FMI and the MONO32 or UHMA models. However, the problem can be solved mathematically, if we use the dispersion model to provide for a more simple function that describes the dilution of the air parcel within a sufficiently good accuracy. We have therefore defined numerical power functions, $y = ax^{-b} = a(ut)^{-b}$, in which the coefficients a and b are positive, for approximating the dilution process, x (in m) refers to the distance from the source and u is the horizontal wind speed perpendicular to the highway. The coefficients a and b are determined separately for each meteorological case, using the CAR-FMI model; this procedure results in an offline type coupling of the dispersion model and the aerosol process model.

The dilution was therefore first computed for an inert pollutant by the FMI-CAR model. The road was assumed to consist of a set of two line sources that were located in the middle of the three lanes, separately for both directions of the traffic. Only the dilution of vehicle exhaust was computed, background was not included in the dilution calculations.

The emission and dilution rates were taken into account in the aerosol process model by appropriately modifying the differential equations of the number and mass concentrations of particles. Dilution rate was obtained by derivating the above mentioned numerical power function as a function of time. To calculate the modal emission rates (particles $\text{cm}^{-3} \text{s}^{-1}$) the modal emission factors Q_i (particles $\text{m}^{-1} \text{s}^{-1}$) have to be divided by the width of three lanes to one direction to obtain the portion of emissions caught by the box, and by the box height since air in the box is assumed to be well mixed.

The time evolution of particle number concentrations (N_i) and the total mass concentration of different compounds j ($=1,2$) ($M_{i,j}$) in i th ($=1,6$) size mode are given by

$$\frac{dN_i}{dt} = -0.5K_{ii}N_i^2 - \sum_{k=i+1}^6 K_{ik}N_iN_k - \lambda_{\text{dep},i}N_i + \frac{Q_i}{Hw} - \frac{b}{t}(N_i - N_{bi}) \quad (1)$$

and

$$\frac{dM_{i,j}}{dt} = N_i I_{i,j} + \sum_{k=1}^{i-1} K_{ik}N_iN_k m_{k,j}$$

$$- \left(\sum_{k=i+1}^6 K_{i,k}N_iN_k \right) m_{ij} - \lambda_{\text{dep},i}N_i m_{i,j} + \frac{Q_i}{Hw} m_{ei,j} - \frac{b}{t} (N_i m_{i,j} - N_{bi} m_{bi,j}) \quad (2)$$

where K_{ik} is the coagulation coefficient, $\lambda_{\text{dep},i}$ is the dry deposition velocity (s^{-1}) of the particles in size class i , H is the box height, $w=12$ m is the width of the lanes to one direction, Q_i is the modal emission factor (in particles $\text{m}^{-1} \text{s}^{-1}$), b is the dilution coefficient, N_{bi} is the number concentration of particles in background air in size class i given by Table 2. $I_{i,j}$ is the mass flux of condensable vapour j (sulphuric acid or organic vapour) to the particles in size class i given by Pohjola et al. (2003). In Eq. (2) $m_{i,1}$ refers to the sulphuric acid mass and $m_{i,2}$ to the organic mass in a particle in size class i ; subscripts b and e refer to the urban background and emitted exhaust particles, respectively. Due to different origin the background and exhaust particles are expected to have different composition.

One additional procedure is needed for integrating the emission modelling (in terms of the total emissions from the traffic flow) and the combined dispersion and aerosol dynamics modelling. The combined dispersion and aerosol process model addresses expanding air parcels that are transported along with the air flow over the road. For simplicity, we address here only cases, in which the wind direction is perpendicular to the road. First, one needs to assume an initial height of the air parcel volume, which corresponds to the situation where vehicular exhaust gases and particles have been diluted in a time scale of less than 0.5 s, after their discharge from the tailpipe. Due to the rapid temperature decrease immediately after the exhaust, nucleation has already occurred within this time-scale (Rönkkö et al., 2006); we have therefore assumed that the diameter of the smallest size mode was 4.7 nm. The initial dispersion is dependent on both the traffic-induced and atmospheric turbulence. Clearly, the height of the initial mixing volume cannot be smaller than the effective release height of the emissions. We have assumed that this initial air parcel height is equal to 0.80 m.

There is a continuous flux of vehicular emissions to the moving air parcel as long as the parcel is transported over the lanes; this is also the case for the modelling. The height of the parcel was assumed to increase linearly above the traffic lanes, in order to account for the effect of dilution during transport above the lane. For this linear dependence we have performed a semi-empirical fitting in the following way. The slope of the linear increase of the height of the air parcel was adjusted so that the total particle number concentration based on one of the 14 dispersion cases (case 10: 19 February at 14:00) agreed with the measured value, which was $1.83 \times 10^5 \text{ cm}^{-3}$ with $\text{SD} = 8.98 \times 10^4 \text{ cm}^{-3}$ (Pirjola et al., 2006). This numerical fit has been presented in Fig. 5. This height increases linearly above the lanes; this takes into account atmospheric dispersion. During the grass area between the lanes as well as downwind from the road, dilution is

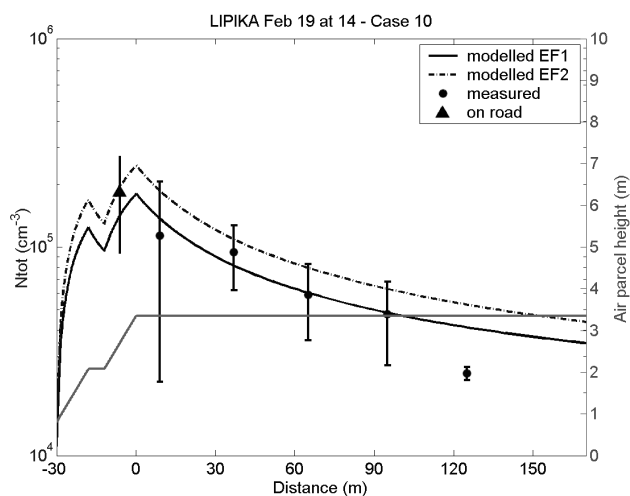


Fig. 5. Measured and predicted results for case number 10 at 02:00 p.m. on 19 February 2003 (MONO32). The predicted results were computed using two sets of emission factors: *EF1* refers to those adopted from Gidhagen et al. (2004) whereas *EF2* refers to those from Yli-Tuomi et al. (2004). The measured concentrations on road (adopted from Pirjola et al., 2006) and in the roadside are shown as a black triangle and black circles, respectively, and the vertical bars show their standard deviations. The semi-empirical fitting of the air parcel height was made according to this case; the figure therefore also shows the air parcel height. See details in the text.

modelled using the CAR-FMI model and the height of the air parcel is therefore constant.

This procedure resulted in a slope of 0.17 m s^{-1} and this value was applied to all the simulations. The x-axis in Fig. 5 is the distance from the roadside instead of time, and since the slope of the air parcel height also depends on wind speed (1.6 m s^{-1} in this case), the slope in the figure is 0.106. The height of the air parcel is assumed to be constant while being transported over the central grass area of the road and after it has crossed the road; dilution and mixing with the background air are subsequently taken into account as described above. As a result, above the downtown direction the box height was increased from 80 cm to 1.56–2.84 m depending on the wind speed ($2.7\text{--}1.0 \text{ m s}^{-1}$), and during the eastern direction to 2.31–4.88 m, respectively.

In the model, particle growth was assumed to occur by condensation of H_2SO_4 and an organic vapour (Rönkkö et al., 2006); however, this vapour is not yet identified and its concentration and thermodynamic properties are therefore not well known. H_2SO_4 is formed photochemically from its precursor SO_2 whose concentration in the background air was assumed to be 1 ppb. The concentration of SO_2 in the exhaust gas was assumed to be $\sim 5.5 \times 10^{11} \text{ molecules cm}^{-3}$ ($\sim 20 \text{ ppb}$), in mega cities it might be even several hundred ppb (Seinfeld and Pandis, 1998), Condensable organic vapour concen-

tration at the roadside was an unknown parameter, and therefore it was varied from 10^9 to $10^{12} \text{ molecules cm}^{-3}$. Hellén et al. (2006) have shown that the average winter concentrations of different VOC compounds at an urban site in Helsinki are $\sim 4.9 \times 10^4 \text{ ng m}^{-3}$ corresponding to $2.9 \times 10^{11} \text{ molecules cm}^{-3}$. We assumed that 10% of the VOC's react to form oxidation products that are condensable (C_{org}). The urban background condensable VOC oxidation product concentration $C_{\text{org,bg}}$ was assumed to be $2.5 \times 10^8 \text{ molecules cm}^{-3}$ ($\sim 10 \text{ ppt}$). SO_2 and organic vapour were diluted in the similar way as particles.

4 Results and discussion

The meteorological data used was from the period 17–20 February 2003, at time periods when the wind was directed from the road towards the measurement sites (total of 14 h with these conditions). Three example cases were chosen for a more detailed examination. These specific cases were selected in order to be able to address various traffic situations, prevailing wind speeds and atmospheric stratifications. The cases are: Case 1 on 17 February at 06:00–07:00 p.m. (Monday, evening hour after busiest rush hours, lower wind speed of 1.3 m s^{-1} , neutral stratification), Case 8 on 19 February at 08:00–09:00 a.m. (Wednesday, morning rush hour, higher wind speed of 2.7 m s^{-1} , neutral stratification) and Case 11 on 19 February at 04:00–05:00 p.m. (Wednesday, afternoon rush hour traffic, lower wind speed of 1.3 m s^{-1} and stable stratification).

4.1 Modelled and measured total number concentrations

The scatter plot of the measured and predicted hourly total number concentrations (at 14 hourly cases) at all roadside measurement points (5–9 locations) is presented in Fig. 6. The overall average Index of Agreement (which is a measure of correlation of two sets of data) between the measured and predicted values of N_{tot} was 0.69. When looking at each day separately, IA varied from 0.96 on 17 February (2 cases) to 0.42 (4 cases) on 20 February while the Fractional Bias (a measure of the agreement of mean concentrations) varied between 1.02 on 17 February and -1.02 on 20 February.

Measured and modelled total number concentrations of particles as a function of distance from the edge of the road for the three example cases are presented in Figs. 7–9a. The measured total number concentration is calculated as the sum of particles smaller than 50 nm recorded by SMPS and particles larger than 50 nm recorded by ELPI (see details in Pirjola et al., 2006). As an example in the 17 February case, presented in Fig. 7a), the measured total number concentration decreased by 43% between the closest measurement point at 9 m distance and 65 m distance from the edge of the road, and by 58% between the distances of 37 m and 95 m from the edge of the road. Number concentration in the nucleation

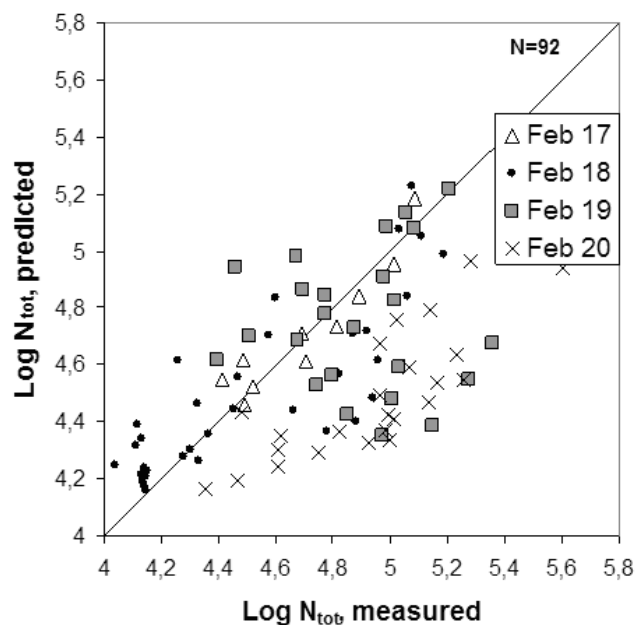


Fig. 6. Predicted (MONO32) total number concentrations versus measured total number concentrations for all studied cases at all roadside measurement locations. The legend shows the measurement days.

1 and 2 and Aitken modes decreased by 40–54% between the closest measurement point at 9 m distance and 65 m distance from the edge of the road, and by 55–60% in the nucleation modes and 36% in the Aitken mode between the distances of 37 m and 95 m from the edge of the road. In the accumulation modes, the modewise number concentration decrease between the distances of 9 m and 65 m was 18–20%, and between the distances of 37 m and 95 m the decrease was 14% for the accumulation 1 mode while the accumulation 2 mode number concentration increased by 1%.

Clearly, such results are also dependent on atmospheric conditions, and could therefore be different, e.g., for various climatic regions and the seasons of the year. However, these results are similar to the findings of Zhu et al. (2002b) and Hitchins et al. (2000), who found that total number concentration decreased to approximately half of its original value at a distance of 30 m at somewhere between the distances of 90 and 150 m. They also found that the number concentration of small particles decreased more rapidly than that for large particles with increasing distance from a freeway.

In all cases, the total number concentrations are in maximum at the roadside ~35% higher by using the emission factors calculated based on the results by Yli-Tuomi et al. (2004) than by Gidhagen et al. (2004). As the distance from the road increases up to 170 m the difference decreases to 25–27%. Neither of the emission factors by Yli-Tuomi et al. (2004) or by Gidhagen et al. (2004) produced consistently better results in comparison with the measured data.

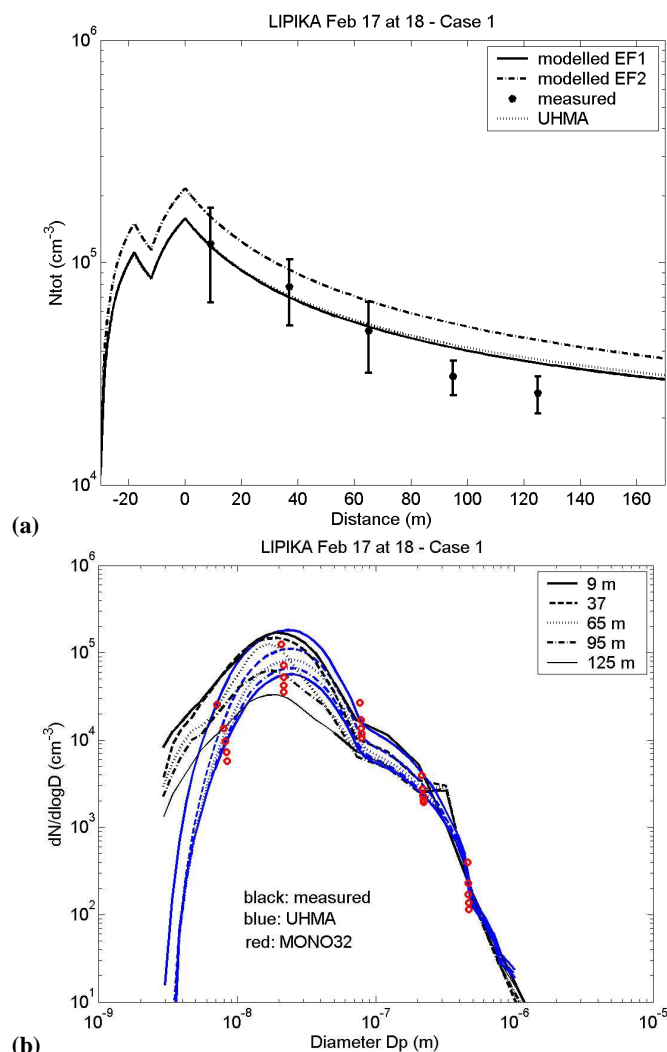


Fig. 7. Measured and predicted results at 06:00–07:00 p.m. on 17 February 2003. **(a)** Measured and modelled total number concentrations of particles as a function of distance from the edge of the road. **(b)** Predicted and measured number concentration distributions at various measurement distances from the road. The five curves in blue, and the five red dots for each diameter correspond to the same distance values as the measured data (from 9 to 125 m).

Also shown in Fig. 7a is the total number concentration predicted by UHMA for Case 1 using the emission factors based on Gidhagen et al. (2004) and the value of $1 \times 10^{10} \text{ cm}^{-3}$ for the condensable insoluble organic vapour. As expected, the time evolutions of the total number concentration computed by the UHMA and MONO32 models almost overlap; at the distance of 150 m from the road the difference of N_{tot} was 4.2%. The slight deviation might be due to the somewhat different subroutine used for calculating the water content of particles.

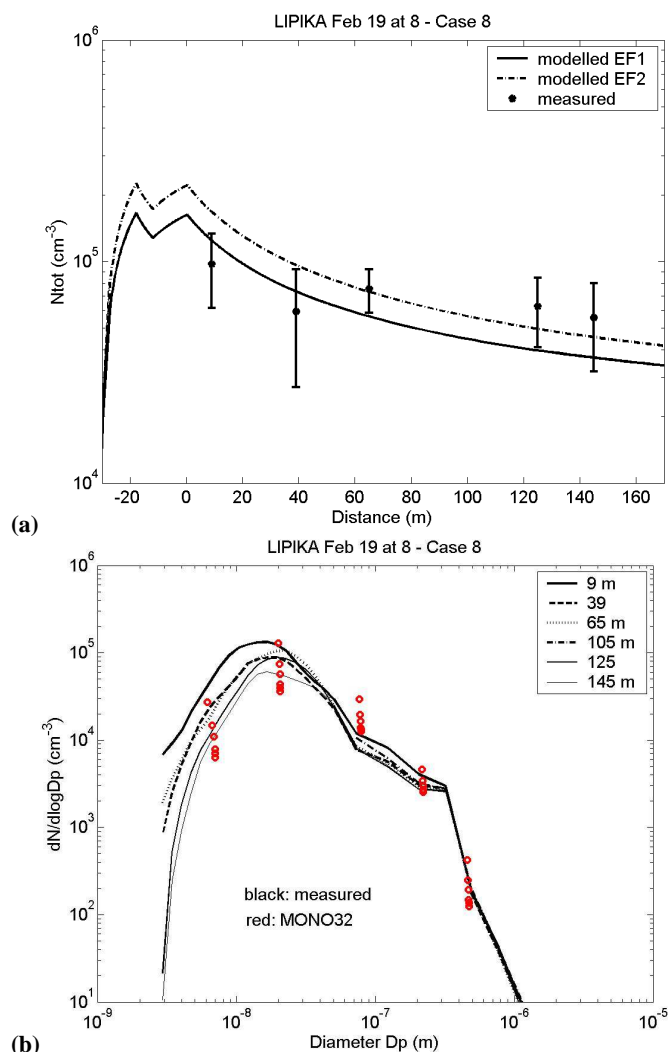


Fig. 8. Measured and predicted results at 08:00–09:00 a.m. on 18 February 2003. **(a)** Measured and modelled total number concentration of particles as a function of distance from the edge of the road. **(b)** Predicted and measured number concentrations distributions at various measurement distances from the road.

In the two other studied cases, presented in Figs. 8a and 9a, either the traffic situation or background changed markedly during the hour within which the measurements at different distances from the road were made. In Case 11 (at 04:00–05:00 p.m. on 19 February 2003), the total number concentration at the edge of the road was the highest, mainly due to the highest traffic density (busy evening traffic) and the fact that there was more traffic on the lane closest to the measurement sites.

4.2 Modelled and measured number size distributions

Predicted and measured number size distributions at the measurement distances from the road are presented in Figs. 7–9b.

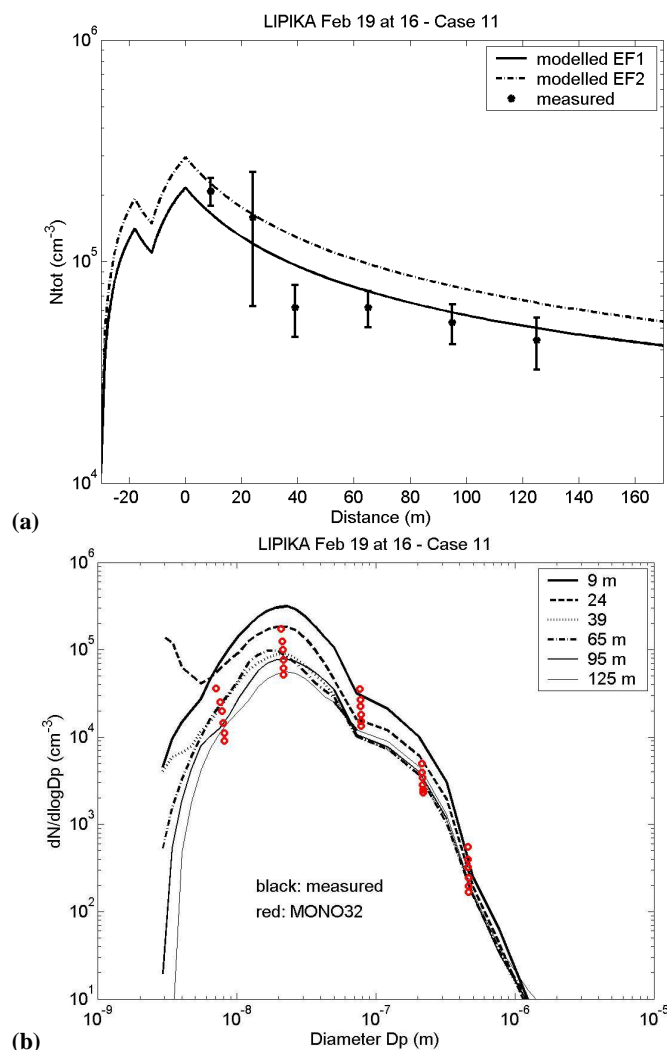


Fig. 9. Measured and predicted results at 04:00–05:00 p.m. on 17 February 2003. **(a)** Measured and modelled total number concentrations of particles as a function of distance from the edge of the road. **(b)** Predicted and measured number concentration distributions at various measurement distances from the road.

The measured results are obtained by SMPS and ELPI. Since in the monodisperse models all particles in each mode (N_i) have the same size which grows as a function of time, they cannot directly describe the lognormal size distribution. Instead, we have used the fixed mode limits given in Fig. 3, and calculated $dN/d\log D_p = N_i / \log(D_p(i+1)/D_p(i))$ in these size sections, $1 \leq i \leq 6$. The base 10 of logarithm was used. For comparison, Fig. 7b also shows the size distribution predicted by UHMA (Case 1).

The size distribution by UHMA, presented in Fig. 7b, agrees well with the measurements; however, there is a slight over-prediction. The results by MONO32 are also in a good agreement with the measurement data, although the

Table 3. The percentual changes of modewise number concentrations and particle diameters from 9 m distance to 125 m distance from the edge of the road in the Case 1, 17 February at 06:00–07:00 p.m. $\Delta X = (X(125\text{ m}) - X(9\text{ m})) / X(9\text{ m}) \times 100$, where $X = \#$ or D . Less or smaller particles at 125 m; $\Delta < 0$, more or larger particles at 125 m; $\Delta > 0$.

Set of C_{org} and processes included			Change of umber concentration in mode: $\Delta\#$						Change in diameter of mode: ΔD_p					
	C_{org}	Processes	Nuc1	Nuc2	Ait	Acc1	Acc2	Coa	Nuc1	Nuc2	Ait	Acc1	Acc2	Coa
Varying C_{org}	1e9	all	−77	−69	−57	−46	−69	−1	4.3	0.9	1.7	3.9	3.3	0.2
	1e 10	all	−76	−69	−57	−46	−69	−1	14	1.9	1.0	4.3	2.9	0
	1e 12	all	−75	−69	−57	−46	−69	−1	190	144	59	13	10	0
Varying set of processes included	1e 10	none	−76	−69	−57	−46	−69	−1	0	0.1	1.6	4.6	3.5	0
	1e 10	cond only	−76	−69	−57	−46	−69	−1	14	1.9	1.0	4.5	3.0	0
	1e 10	coag only	−78	−69	−57	−46	−69	−1	0.4	0.2	1.6	3.6	3.5	0
Measured data			−76	−80	−56	−23	−10	−43	1.7	−1.8	0.1	15	0.3	−7.5

monodisperse models cannot give detailed information of the nucleation mode particles. However, this is also the case for many instruments such as for example, ELPI, which provides only two size sections for particles in the size range of 7–56 nm.

Clearly, based on the results in Fig. 7b for the cases studied, the growth rate of nucleation mode particles is over-predicted by both models. Sensitivity simulations (not shown) indicate that this is most likely caused by the uncertainties associated with the modelling of the condensation of an organic vapour. As the physical and chemical properties of organic vapours in urban air are poorly known, it is not possible to predict accurately condensing vapour fluxes at specific ambient conditions. However, the uncertainties associated with the properties of an organic vapour affect only the sizes of particles that are smaller than approximately 10 nm, and these do not limit our ability to simulate most of the size distribution or the total particle number concentration.

When considering the results of the MONO32 model, during the 17 February case period, the measured N_{tot} was on average (over all the measurement distances) 87% of the modelled N_{tot} values, during the 18 February case period it was 129%, and during the 19 February case period, about 76%. A shift towards larger diameter is seen in the modeled smaller nucleation mode size bin in all three cases, by 14%, 19% and 14% in chronological order of the cases. This shift is less pronounced in the second nucleation bin (2–3%) and the Aitken bin (1–2%), and a bit more pronounced in the two accumulation modes (3.6–4.3% for first accumulation bin and 2.4–3.9% for the second accumulation mode).

Ketzel et al. (2004) found that for a roadside site in Copenhagen, the maximum in size distribution is at about 22 nm in traffic, 20–30 nm at kerbside and 50–60 nm at a rural site. As presented in Fig. 3, the maximum in the size distribution at the Itäväylä site was at 18 nm in traffic, about 19 nm at 9 m distance from the edge of the highway, and 16–17 nm at the background site.

We have also evaluated the evolution of particle size distribution assuming various concentrations of condensable vapours for the 17 February case. These results are presented in the Fig. 10; however, the results for the coarse mode have not been shown, as there is no variation in terms of the concentration of the condensable vapour. The percentual changes of modewise number concentrations and particle diameters from 9 m distance to 125 m distance from the edge of the road are presented in the Table 3. The highest concentration of condensable organic vapour (C_{org}) used, 10^{12} molecules cm^{-3} , was clearly too high, as five of smallest predicted size modes at the distance of 37 m had already grown larger than 10^{-7} m in diameter. For the nucleation 1 mode, the $C_{\text{org}} = 10^9$ molecules cm^{-3} produced diameter values closest to the measured ones, for nucleation 2 mode, the $C_{\text{org}} = 10^{10}$ molecules cm^{-3} was the best, and for the other size bins, the predicted sizes were very similar. Considering all measured data, the changes of the predicted particle number concentrations were of the same order as the corresponding measured values with both values of C_{org} , 10^9 and 10^{10} molecules cm^{-3} in the nucleation and Aitken modes, overestimated by factor of 2 for accumulation 1 mode and by factor of 7 for accumulation 2 mode, and underestimated by factor of 43 for the coarse mode. The order of magnitude of the concentration of condensable organic vapours could therefore be estimated from these results.

The importance of aerosol processes was also evaluated for the 17 February case for the six size bins by modelling runs with all processes included being compared to modelling runs with each aerosol process being excluded from the model equations. As an example, the results considering the particle effects on particle diameter in the case with C_{org} being 10^{10} molecules cm^{-3} are presented on the right side of the Table 3. The effect of condensation is most pronounced for the nucleation 1 mode, where the model runs with no processes or only coagulation included seems to be more accurate. For the nucleation 2 mode, the results with

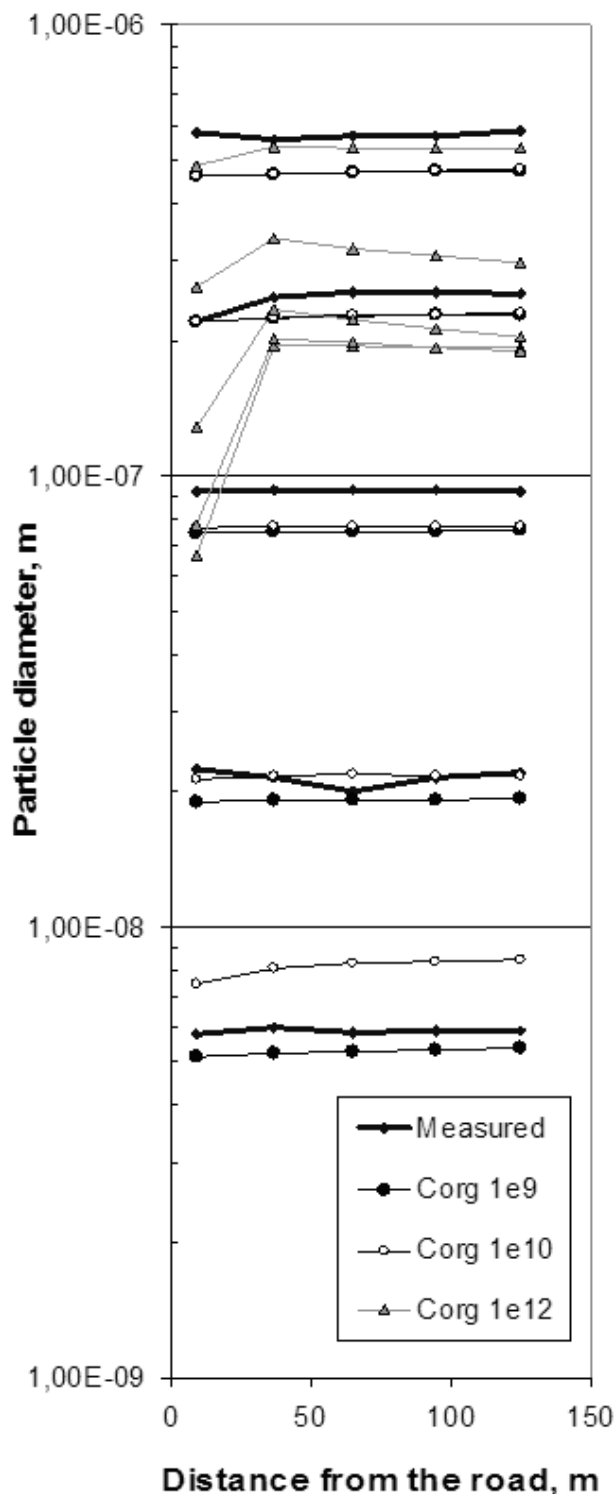


Fig. 10. Measured and modelled particle diameters as a function of the distance from the edge of the road for Case 1 at 06:00–07:00 p.m. on 17 February 2003 (MONO32). The modelled diameters have been computed assuming three different concentrations of the condensable organic vapour.

condensation only included in the model run are more accurate. For the other size bins, the model runs produce similar results whether condensation and coagulation are included or not. This means that in the present conditions, the time scales for self-coagulation of nucleation mode particles as well as inter-modal coagulation of nucleation mode particles with larger particles are longer than the time scale for dilution (see e.g. Kerminen et al., 2007).

The measured decrease of the diameters of the nucleation 2 and coarse mode particles is not reproduced by the modelling. Jacobson and Seinfeld (2004) found that at 30 m downwind, the difference in number concentration between total number concentrations in the no-coagulation and the coagulation case suggested that about 2% of the decrease in particle concentration between 0 and 30 m was due to coagulation and most of the rest was due to dilution (some was due to dry deposition as well). This result, though, depends substantially on the emission rate. In our example case on 17 February, the difference in total number concentration (not presented) in model runs with all aerosol processes included and coagulation excluded, the effect of coagulation was about 1% decrease in particle total number concentration at 37 m distance from the edge of the road. In the model run with condensation excluded the particle number concentration was increased by 0.3% at 37 m distance compared to the run with all aerosol processes included.

4.3 Comparison of modelled values and the background

The total particle number concentration of the urban background used in the modeling was $1.11 \times 10^4 \text{ cm}^{-3}$, while at the farthest modelled distance of 350 m (wind speed in this case was 5.2 m s^{-1} and deviation of wind speed from perpendicular to the highway was about 8°) the total number concentration was still above the background with the value $1.32 \times 10^4 \text{ cm}^{-3}$. The corresponding size number distributions are presented in Fig. 11. The modelled particle mode sizes and mode wise number concentrations are larger (by 6–50% and 4–145%, respective) than the input values with particle sizes smaller than 120 nm, while modelled particles larger than this value were 35% smaller in size and 22% more in number concentration.

Taking the sensitivity to wind speed and the used dilution model into account, this is similar to the results of Zhu et al. (2002a), who found in their study that UF particle number concentration measured at 300 m downwind was indistinguishable from the upwind background number concentration. Zhang and Wexler (2004) also state, based on theoretical analysis, that road plumes become ambientlike in their particulate matter characteristics at about 300 m distance from the road. However, neither of these studies assessed the evolution of the particle diameters in the comparison.

5 Conclusions

In Part I of this study we have used the data of a recently conducted roadside measurement campaign for the evaluation of combined dispersion and aerosol process models. The campaign contained data on particle number concentrations, size distributions and the concentrations of CO and NO_x. The experimental dataset is extensive and versatile both in terms of the measured aerosol quantities and the locations of measurement sites. The data contains both measured particle number concentrations and size distributions at several distances from the road, as well as urban background measurements and number concentration measurements on the road. The main limitations of the experimental campaign are probably associated with the fairly limited temporal duration of the campaign, i.e., two weeks. In addition, there were only four days suitable for modelling of the influence of the emissions on the road (downwind cases on 17–20 February 2003). Clearly, the importance of various aerosol processes depends on the spatial scale; this study addresses the scale that is smaller than 200 m regarding measured data and 360 m concerning the model computations.

In this study, we have compared the measured data with the predictions of the road network dispersion model CAR-FMI used in combination with an aerosol process model MONO32. Compared to previous studies in this area (e.g., Pohjola et al., 2003, 2004), we have utilised a state-of-the-art atmospheric dispersion model and atmospheric boundary layer scaling. It is straightforward to show that there is no analytic mathematical solution for combining the equations of these two models. We have therefore developed an approximative mathematical method that (i) presents the dilution of pollution as simplified equations that are based on the roadside dispersion model, and (ii) combines these equations mathematically with the set of differential equations that constitute the aerosol process model. The same or similar mathematical procedure could probably be used also for coupling other dispersion models representing various spatial scales, and aerosol process models.

In order to be able to inter-compare the predictions of two aerosol process models, we have also used the sectional aerosol process model UHMA for one of the cases. The predictions of both the MONO32 and UHMA models (both combined with the CAR-FMI model) were in a good agreement with the measurements, for this example case. Regarding the application of the models, the probably most uncertain aspect is the assessment of the source term for atmospheric dispersion above the road, including especially the initial air parcel mixing height, and the modelling of its increase over the road.

In this study, three example cases were chosen for a more detailed examination. These specific cases were selected in order to be able to investigate various traffic situations, prevailing wind speeds and atmospheric stratifications. Considering the evolution of total number concentration, dilution

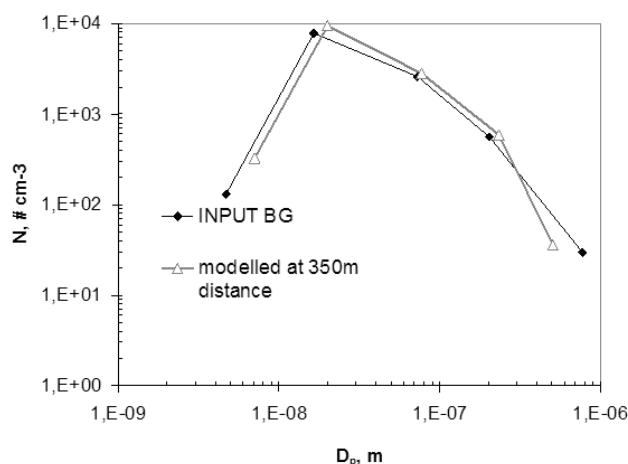


Fig. 11. The particle number size distribution of the urban background (squares) used as input values in the aerosol modelling (Table 2), and the modelled (MONO32) particle number size distribution at the distance of 351 m from the road, both of these on at 10:00–11:00 p.m. on 18 February 2003. The wind speed was 2.7 m s^{-1} and direction $\sim 330^\circ$.

was shown to be the most important process on the distance scale considered. In the cases reported here, the aerosol number concentration is fairly low and the dilution is not in any way inhibited (i.e., its characteristic time scale is relatively short); these conditions are unfavourable especially for coagulation. The rates at which the number concentrations of particles in different size modes decrease as a function of distance from the road were dependent on the urban background aerosol size distribution, as the background to on-road-ratio of aerosol number concentration was dependent on particle size. The influences of coagulation and condensation on the number concentrations of particle size modes were found to be negligible on this distance scale. Condensation was found to affect the evolution of particle diameter in the two smallest particle modes. The assumed value of the concentration of condensable organic vapour of 10^9 – $10^{10} \text{ molecules cm}^{-3}$ resulted in particle sizes that were closest to the measured values.

The particle number size distribution of the plume originated from the traffic emissions in the major road studied approached closely that of the urban background at a distance of approximately 300 m. Clearly, this result depends on the magnitude of the urban background, the traffic density on the road studied and the dispersion conditions. However, the result obtained in this study was very similar to the previous estimates by Zhu et al. (2002a) and Zhang and Wexler (2004).

The atmospheric stability changed during the four measurement campaign days, and the conditions grew more stable towards the end of the period. For the most stable cases, the simulation results were relatively less accurate. The

sensitivity of the performance of the CAR-FMI model in terms of atmospheric stability has been studied against a substantially more extensive dataset in a previous study (Kousa et al., 2001). In that study, it was found out that the modelling system tends to slightly underpredict the measured concentrations in convective atmospheric conditions, and overpredict in stable conditions. During stable atmospheric stratification, the relatively slower mixing with background air provides additional time for condensation and coagulation to proceed, as was also found out by Kerminen et al. (2007).

A specific limitation of the campaign is that both the measurements of the particle size distributions and those of the total number concentrations (N_{tot}) at various distances from the road are not simultaneous, but from subsequent time periods, as only one measurement van was available for the campaign. The concentrations at different distances were measured during approximately 5–10 min of parking at that particular location, and the measurements were started at the site that is closest to the edge of the road. This causes a stochastic variation to the measured concentration data, when the data at various distances from the road are inter-compared. Clearly, the measured concentrations can therefore randomly vary in terms of the short-term variations of the traffic densities, driving speeds and meteorological conditions, while the modeling results are based on hourly averaged input values of the meteorological and traffic flow data. The commonly occurring short-term variation of the measured concentrations at the stationary measurement site (the site at the distance of 9 m from the edge of the road) was about 30% during one measuring cycle of approximately one hour (Pirjola et al., 2004).

In Part I of this study, we have focused on the modelling of the particle size distributions and number concentrations as a function of distance from the road; however, in Part II, modelling has only a supplementary role. Part II of this study focuses on the evaluation of the temporal variations and dependencies on local meteorological conditions of the measured particle number concentrations and size distributions at one distance from the road, and compares these with the corresponding measured data at an urban background location.

Regarding the experimental data, these two papers (Part I and II) share one common location in the vicinity of the Itäväylä road. In Part I, the background concentration data measured at the station of Kumpula was not used; however, this was an essential component of the studies of Part II. The time periods studied were different, February of 2003 in case of Part I, and from August to September of 2003 and from January to February of 2004 in case of Part II. Consequently, the prevailing meteorological conditions were substantially different for these two (so-called LIPIKA and SAPPHIRE) campaigns; the meteorological variability contributed to the differences of the vehicular emissions and the background concentrations (regarding both the size distributions and the number concentrations).

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